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Structure and Magnetic Properties of $Sm(Co_{bal}Fe_xTi_{0.05})_{9.66}$ Alloys (x = 0–0.57) and $Sm(Co_{0.66}Fe_{0.19}Ti_{0.05}Cu_{0.1})_{9.66}$ Magnets

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***Abstract*—Potential permanent magnetic materials with compositions of $Sm(Co_{bal}Fe_xTi_{0.05})_{9.66}$ (x = 0–0.57) have been synthesized and characterized in the temperature range of 10–1473 K and at fields up to 5 T. The experimental results show that near single phase materials with Th_2Ni_{17} structure were formed in $Sm(Co_{bal}Fe_xTi_{0.05})_{9.66}$ alloys after splat quenching from 1473 K. The Ti atoms play an important role in stabilizing the Th_2Ni_{17} structure for the 3d (transition metal) rich nonstoichiometric 2–17 compounds. Encouraging hard magnetic properties with $T_c \sim$ 890–1066 K, $M_s \sim$ 10.8–13.7 kG, $H_a \sim$ 30–125 kOe at 300 K were observed in $Sm(Co_{bal}Fe_xTi_{0.05})_{9.66}$ alloys. Both $Sm(Co_{0.66}Fe_{0.19}Ti_{0.05}Cu_{0.1})_{9.66}$ sintered and melt-spun powder magnets with $4\pi M_s \geq$ 10 kG were fabricated. A strong domain wall pinning behavior with $H_c \sim$ 1.6 kOe at RT and $H_c \sim$ 4.3 kOe at 10 K was observed. The effect of different heat treatment conditions on the phase formation of $Sm(Co_{bal}Fe_xTi_{0.05})_{9.66}$ alloys was also discussed.**

***Index Terms*—3-D rich magnetic materials, Curie temperature, Th_2Ni_{17} structure, uniaxial anisotropy.**

I. Introduction

The coercivities of Sm–Co 2–17 type magnets at high temperature have been significantly improved by reducing the 3d/R ratio down to 7 [1], [2], at the expense, however, of their high magnetic moments. In such a context, further increase in saturation magnetization without reductions in coercivities becomes the next challenging goal. One of our efforts is to search for new compounds with much higher transition metal content ($3d/R > 8.5$), high Curie temperature T_c, and high anisotropy H_a. Our previous work on $(Sm_xPr_{1-x})_3$ $Fe_{27.5}Ti_{1.5}$ alloys (x = 0–1) with $3d/R \sim 9.67$ [3] has been expended to compositions on the Co-rich side. This paper reports potential permanent magnetic materials with compositions of $Sm(Co_{bal}Fe_xTi_{0.05})_{9.66}$ (x = 0–0.57). The magnetic properties of both $Sm(Co_{0.66}Fe_{0.19}Ti_{0.05}Cu_{0.1})_{9.66}$ sintered and melt-spun powder magnets are also presented.

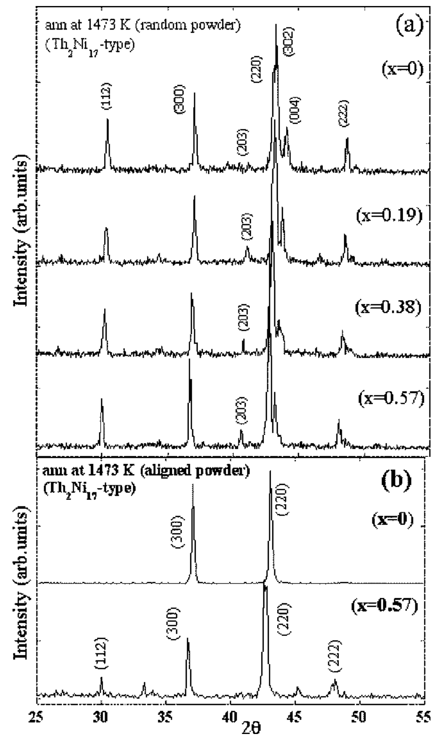

Fig. 1. XRD of (a) random powder and (b) aligned powder of $Sm(Co_{bal}Fe_xTi_{0.05})_{9.66}$ alloys (x = 0–0.57) after splat cooling from 1473 K.

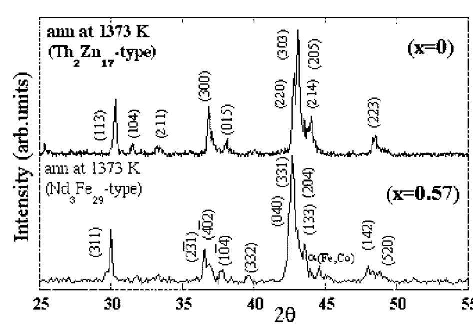

Fig. 2. XRD of $Sm(Co_{bal}Fe_xTi_{0.05})_{9.66}$ alloys (x = 0 and 0.57) after annealing at 1373 K.

II. Experimental Details

The alloys were prepared by induction melting under Ar atmosphere, and subsequently annealing at 1323 K–1473 K for

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TABLE I
LATTICE PARAMETERS AND MAGNETIC PROPERTIES OF $Sm(Co_{bal}Fe_xTi_{0.05})_{9.66}$ ALLOYS ($x = 0$–0.57) (AFTER SPLAT COOLING FROM 1473 K, FORMED Th_2Ni_{17} STRUCTURE)

x	a (Å)	c (Å)	c/a	Ms(300K) (kG)	Ms(10K) (kG)	Ha(300K) (kOe)	Ha(10K) (kOe)	Tc(K)	$(BH)_{max}$ theo. (MGOe)
0	8.399	8.210	0.977	10.8	11.6	125	150	1066	29
0.19	8.410	8.288	0.985	12.0	12.7	100	140	1053	36
0.38	8.437	8.331	0.987	13.6	14.0	70	95	995	46
0.57	8.479	8.375	0.988	13.7	14.6	30	50	890	47

24 h. X-ray diffraction (XRD) with Cu radiation was used to determine the crystal structure and phases present. Magnetic properties (M_s, T_c, H_a, and H_c) were measured in the temperature range of 10 K–1473 K and at fields up to 5 T, by using a vibrating sample magnetometer (VSM) and a superconducting quantum interference device (SQUID) magnetometer. Samples were in the forms of chunk, loose or aligned powder (<38 μm). The anisotropy field (H_a) was estimated by extrapolation of the difference between the easy axis M and hard axis M to zero. Honda extrapolations were utilized to determine the saturation magnetization M_s. An alloy with composition of $Sm(Co_{0.66}Fe_{0.19}Ti_{0.05}Cu_{0.1})_{9.66}$ was processed into magnets by the conventional powder metallurgy and melt-spun powder technique. The BH loops of sintered magnets were measured by a BH loop tracer.

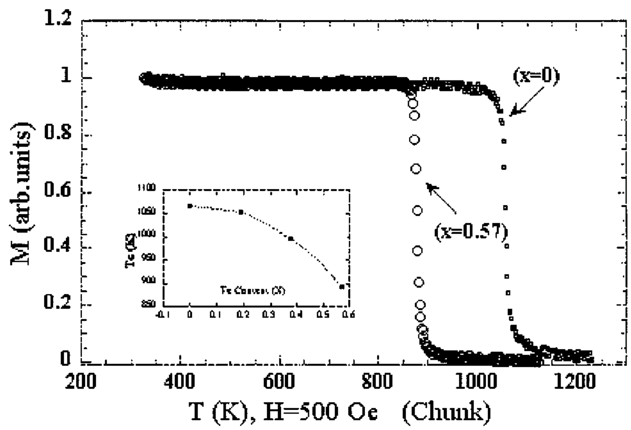

Fig. 3. M versus T of $Sm(Co_{bal}Fe_xTi_{0.05})_{9.66}$ alloys ($x = 0$ and 0.57) after splat cooling from 1473 K and Curie temperatures Tc versus Fe content x of $Sm(Co_{bal}Fe_xTi_{0.05})_{9.66}$ alloys (insert).

III. RESULTS AND DISCUSSIONS

A. Phases Formed and Structure

Information regarding the structure and phases present is shown in Figs. 1, 2, and Table I.

As shown in Fig. 1(a), the XRD patterns indicate that near single phase materials with a disordered hexagonal Th_2Ni_{17} structure are observed in all $Sm(Co_{bal}Fe_xTi_{0.05})_{9.66}$ alloys ($x = 0$–0.57) after splat cooling from 1473 K. Ti atoms and/or the heat treatment process play important roles in stabilizing the Th_2Ni_{17} structure for these 3d-rich nonstoichiometric 2–17 compounds. The lattice constants of Fe-free sample are estimated to be $a = 8.399$ Å, $c = 8.210$ Å and $c/a = 0.977$. The unit cell appears to be slightly larger than that of Ti free stoichiometric Sm_2Co_{17} ($a = 8.373$ Å, $c = 8.165$ Å) [4]. It can be seen in Table I that the lattice constants a, c and c/a of $Sm(Co_{bal}Fe_xTi_{0.05})_{9.66}$ alloys increase linearly with the Fe content x. It was also found that the structure and phases formed in the $Sm(Co_{bal}Fe_xTi_{0.05})_{9.66}$ alloys vary significantly with composition and heat treatment conditions. When annealed at lower temperatures (1323 K–1373 K), instead of forming the Th_2Ni_{17}-type phase, other magnetic phases were formed in $Sm(Co_{bal}Fe_xTi_{0.05})_{9.66}$ alloys. Specifically, as shown in Fig. 2, for the alloys with $x = 0$–0.38, the other 2–17 phase with a rhombohedral Th_2Zn_{17}-type structure was formed. For the alloy with $x = 0.57$, however, the main phase became a 3–29 phase with a monoclinic Nd_3Fe_{29}-type structure [3].

From the XRD patterns on magnetically aligned powders of $Sm(Co_{bal}Fe_xTi_{0.05})_{9.66}$ alloys, one can easily detect their magnetic anisotropy behavior. As seen in Fig. 1(b), two strengthened lines, (300) and (220), indicate that all of the alloy samples ($x = 0$–0.57) exhibit a strong uniaxial anisotropy after quenching from 1473 K. The intensity of anisotropy decreases slightly as the Fe content increased. The uniaxial anisotropy behavior was also observed in the alloy samples ($x = 0$–0.38) with Th_2Zn_{17} structure after annealing at 1373 K. For the alloy sample ($x = 0.57$) with Nd_3Fe_{29} type structure, a conical anisotropy was observed. The previous anisotropy behaviors were similar to those of the stoichiometric 2–17 or 3–29 type R–3d compounds [3], [5].

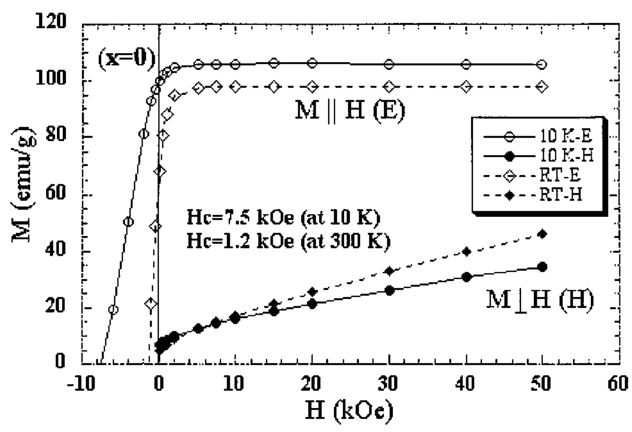

Fig. 4. M versus H (at 300 K and 10 K) of $Sm(Co_{0.95}Ti_{0.05})_{9.66}$ alloy.

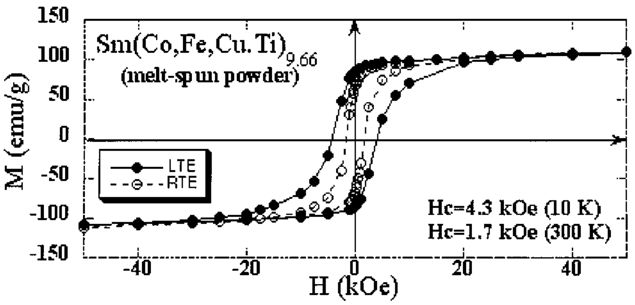

Fig. 5. M–H loops (at 300 and 10 K) of $\mathrm{Sm(Co_{bal}Fe_{0.19}Cu_{0.1}Ti_{0.05})_{9.66}}$ melt-spun powder magnet.

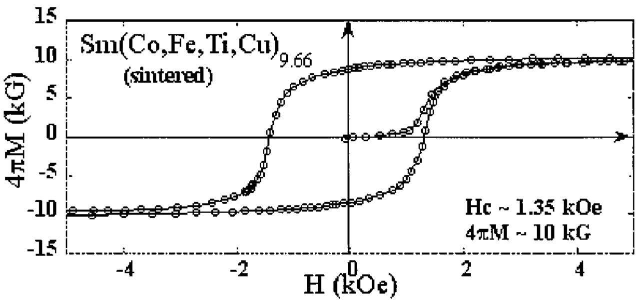

Fig. 6. 4π M–H loop (at 300 K) of $\mathrm{Sm(Co_{bal}Fe_{0.19}Cu_{0.1}Ti_{0.05})_{9.66}}$ sintered magnet.

B. Magnetic Properties

As listed in Table I, and illustrated in Figs. 3–6, encouraging hard magnetic properties with a strong anisotropy $H_a \sim$ 30–125 kOe at 300 K and 50–150 kOe at 10 K; $T_c \sim$ 890–1066 K, and magnetic moment $M_s \sim$ 10.8–13.7 kG at 300 K in the $\mathrm{Sm(Co_{bal}Fe_{x}Ti_{0.05})_{9.66}}$ alloys after annealing at 1473 K were observed. The $\mathrm{Sm(Co_{0.95}Ti_{0.05})_{9.66}}$ possesses a much higher H_a (125 kOe versus 65 kOe [5]) and comparable M_s when compared to that of $\mathrm{Sm_2Co_{17}}$ [5], [6]. As expected, with increasing Fe content, the anisotropy was slightly reduced. However, the saturation magnetic moment was further increased and the estimated theoretical maximum energy products $(BH)_{\max}$ could reach to 46 MGOe at 300 K for the alloy with $T_c \sim$ 995 K. The $\mathrm{Sm(Co_{bal}Fe_{x}Ti_{0.05})_{9.66}}$ alloys could be a potential permanent-magnet materials for high temperature application.

Without Ti addition, as expected, the $\mathrm{SmCo_{9.66}}$ alloy is a mixture of $\mathrm{Sm_2Co_{17}}$ and α–Co phases after annealed at 1473 K via XRD and TMA. However, for the alloys with Ti addition, after annealed at 1473 K, only one magnetic phase with the $\mathrm{Th_2Ni_{17}}$ structure could be detected by XRD and TMA, and no 3d phase(s), such as Co, Co–Ti, Co–Fe or Co–Fe–Ti, were detected. Question arises concerning the $\mathrm{Sm(Co_{bal}Fe_{x}Ti_{0.05})_{9.66}}$ alloys as to where the extra 3d atoms (Co, Fe, and Ti) are located in the $\mathrm{Th_2Ni_{17}}$ unit cell? One possibility would be that some of the Sm atomic sites, 2b or 2d in the $\mathrm{Th_2Ni_{17}}$ unit cell, are partly occupied by the extra 3d atoms. Further investigation via neutron diffraction would be needed to explore this issue.

Both $\mathrm{Sm(Co_{bal}Fe_{0.19}Ti_{0.05}Cu_{0.1})_{9.66}}$ sintered and melt-spun powder magnets with $4\pi M >$ 10 kG, $H_c \sim$ 1.4–1.7 kOe at 300 K and 4.3 kOe at 10 K were fabricated. As seen in Figs. 5 and 6, strong domain wall pinning behavior was observed in the sintered magnets. The H_c could be further enhanced by strengthening the pinning force.

IV. Conclusion

$\mathrm{Sm(Co_{bal}Fe_{x}Ti_{0.05})_{9.66}}$ (x = 0–0.57) alloys can be formed as near single phase materials with $\mathrm{Th_2Ni_{17}}$ structure after splat quenching from 1473 K. The Ti atoms play an important role in stabilizing the $\mathrm{Th_2Ni_{17}}$ structure. The $\mathrm{Sm(Co_{bal}Fe_{x}Ti_{0.05})_{9.66}}$ alloys, with their attractive hard magnetic properties could be a potential permanent magnetic materials for high temperature applications.

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